



UNIVERSITI PUTRA MALAYSIA

***OPTIMIZATION OF PROCESS CONDITIONS AND EMULSION
COMPOSITION, AND THEIR EFFECTS ON EMULSION AND
ENCAPSULATION PROPERTIES OF SPRAY-DRIED FISH OIL POWDER***

MINA TIRGAR

FSTM 2012 11

**OPTIMIZATION OF PROCESS CONDITIONS AND EMULSION
COMPOSITION, AND THEIR EFFECTS ON EMULSION AND
ENCAPSULATION PROPERTIES OF SPRAY-DRIED FISH OIL POWDER**

By

MINA TIRGAR

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in
Fulfilment of the Requirements for the Degree of Master of Science**

June 2012

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

OPTIMIZATION OF PROCESS CONDITIONS AND EMULSION COMPOSITION, AND THEIR EFFECTS ON EMULSION AND ENCAPSULATION PROPERTIES OF SPRAY-DRIED FISH OIL POWDER

By

MINA TIRGAR

June 2012

Chairman : Prof. Jinap Selamat, PhD

Faculty : Food Science and Technology

Fish oil is the richest dietary source of long-chain polyunsaturated fatty acids (LC-PUFAs), but it is prone to oxidative deterioration when exposed to oxygen. The purpose of this study was to produce microencapsulated fish oil powder using spray-drying technology. This study was performed in order to 1) screen the most suitable coating material and optimize the best emulsion formulation for the production of microencapsulated fish oil powder and 2) optimize the spray-drying parameters to produce microencapsulated fish oil powder with the best encapsulation properties. To meet the first objective, three coating materials were used: maltodextrin (15, 25% w/w), Arabic gum (2.5, 7.5% w/w), and methylcellulose (0.5, 1.5% w/w). In addition, the emulsion composition was optimized according to two factors: the fish oil content (6-12% w/w) and the coating material (15–31% w/w) using response surface methodology

(RSM). The results indicated that Arabic gum (5% w/w) had the most significant ($p < 0.05$) effect on the surface mean diameter of the emulsion. Maltodextrin had the most significant ($p < 0.05$) effect on the centrifugal stability of the emulsion and the amount of surface oil of the powder at 15 and 20% (w/w) respectively, whereas methylcellulose (0.5% w/w) had the most significant ($p < 0.05$) effect on the width distribution of the droplets in the emulsion. The oil content (11% w/w) had the most significant ($p < 0.05$) effect on the centrifugal stability and the creaming stability of the emulsion. In addition, oil content (9% w/w) had the most significant effect on the moisture content of the powders. Whereas the interaction between the coating and the oil had the most significant ($p < 0.05$) effects on the surface mean diameter of the powder.

In the second objective, the temperature (140-200 °C), pressure (1-5 kgf/cm²) and feed flow rate of the emulsion (5-20 ml/min) were applied to optimize the spray-drying parameters using RSM. The results indicated that pressure had the most decreasing effect on the average particle size and the amount of surface oil at 4 and 3 kgf/cm² respectively. Moreover, pressure (4 kgf/cm²) had the most increasing effect on microencapsulation efficiency, and the interaction between this factor and the temperature revealed the most significant ($p < 0.05$) effect on total oil extraction. The flow rate had the most significant ($p < 0.05$) effect on the uniformity and color at 7 and 17 ml/min, respectively. No significant ($p > 0.05$) differences were observed between the experimental and predicted values; this verified that the used model for this study was suitable.

A fish oil emulsion containing 17.30% (w/w) coating material consisting of 11.85% (w/w) maltodextrin, 4.80% (w/w) Arabic gum, and 0.65% (w/w) methylcellulose with 11.48% (w/w) oil content and 71.22% (w/w) deionized water was predicted to produce the best coating material and emulsion composition for the encapsulation of fish oil powder. An inlet temperature of 153 °C, pressure of 4 kgf/cm² and feed flow rate of emulsion of 17 ml/min were demonstrated to be the optimum conditions for producing encapsulated fish oil powder.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

**PENGOPTIMUMAN DARI SYARAT PROSES DAN KOMPOSISI EMULSI,
DAN KESANNYA KE ATAS CIRI-CIRI EMULSI DAN PENGKAPSULAN
SEMBURAN-KERING SERBUK MINYAK IKAN**

Oleh

MINA TIRGAR

Jun 2012

Pengerusi : Prof. Jinap Selamat, PhD

Fakulti : Sains dan Teknologi Makanan

Minyak ikan adalah sumber diet terkaya bagi rantai panjang asid lemak politaktepu (LC-PUFA), tetapi ia cepat mengalami kemerosotan oksidatif apabila terdedah kepada oksigen. Tujuan kajian ini adalah untuk menghasilkan serbuk minyak ikan terkapsul mikro dengan menggunakan teknologi pengeringan semburan. Kajian ini telah dilakukan untuk: 1) menyaring bahan salutan yang paling sesuai dan mengoptimumkan formulasi emulsi terbaik untuk pengeluaran serbuk minyak ikan terkapsul dan 2) untuk mengoptimumkan parameter pengeringan semburan untuk menghasilkan serbuk minyak ikan terkapsul dengan sifat pengkapsulan terbaik. Untuk memenuhi objektif pertama, tiga bahan salutan telah digunakan: maltodekstrin (15, 25% w/w), gum arab (2.5, 7.5% w/w), dan metilselulosa (0.5, 1.5% w/w). Disamping itu, komposisi emulsi telah dioptimumkan menggunakan dua faktor: kandungan minyak ikan (6-12% w/w) dan

kandungan bahan salutan (15-31% w/w) dengan menggunakan kaedah gerak balas permukaan (RSM). Keputusan menunjukkan gum arab (5% w/w) mempunyai kesan yang paling ketara ($p < 0.05$) pada purata garis pusat permukaan emulsi. Maltodekstrin mempunyai kesan yang paling ketara ($p < 0.05$) terhadap kestabilan empar emulsi dan jumlah permukaan minyak serbuk pada 15 dan 20% w/w masing-masing, manakala metilselulosa (0.5% w/w) menunjukkan kesan yang paling ketara ($p < 0.05$) ke atas taburan lebar titisan dalam emulsi. Kandungan minyak (11% w/w) mempunyai kesan yang paling ketara ($p < 0.05$) terhadap kestabilan empar dan kestabilan mengkrim emulsi. Di samping itu, kandungan minyak (9% w/w) mempunyai kesan yang ketara ke atas kandungan lembapan serbuk. Manakala interaksi antara lapisan dan minyak mempunyai kesan paling ketara ($p < 0.05$) ke atas purata garis pusat permukaan serbuk.

Dalam objektif kedua, suhu (140-200 °C), tekanan (1-5 kgf/cm²) dan kadar suapan aliran emulsi (5-20 ml/min) telah digunakan untuk mendapatkan parameter optimum bagi pengeringan semburan dengan menggunakan RSM. Hasil kajian menunjukkan tekanan mempunyai kesan paling kurang ke atas purata saiz zarah dan jumlah permukaan minyak pada 4 dan 3 kgf/cm² masing-masing. Tambahan pula, tekanan (4 kgf/cm²) mempunyai kesan yang paling meningkat pada kecekapan pemikrokapsulan, dan interaksi antara faktor ini dan suhu memberi kesan yang paling ketara ($p < 0.05$) pada jumlah minyak terekstrak. Kadar aliran mempunyai kesan yang paling ketara ($p < 0.05$) pada keseragaman dan warna pada 7 dan 17 ml/min, masing-masing. Tiada perbezaan yang ketara ($p > 0.05$) telah diperhatikan di antara nilai eksperimen dan ramalan; ini mengesahkan model yang digunakan dalam kajian ini adalah sesuai.

Emulsi minyak ikan yang mengandung 17.30% (w/w) bahan salutan yang terdiri daripada 11.85% (w/w) maltodekstrin, 4.80% (w/w) gum arab, dan 0.65% (w/w) metilselulosa serta 11.48% (w/w) kandungan minyak dan 71.22% (w/w) air ternyahion telah diramalkan untuk menghasilkan bahan salutan dan komposisi emulsi yang terbaik untuk serbuk minyak ikan terkapsul. Suhu masuk 153 °C, tekanan 4 kgf/cm² dan kadar suapan aliran emulsi sebanyak 17 ml/min telah menunjukkan keadaan optimum untuk menghasilkan serbuk minyak ikan terkapsul.

ACKNOWLEDGEMENT

First of all, I thank God for allowing me to complete my MSc thesis. I am honored and grateful to have the full support from a number of persons in my life for their commitment during my Master's degree.

I would like to express my sincere appreciation and gratitude to Professor Dr. Jinap Selamat for her kindness, support and guidance in all throughout my MSc program.

Special thanks are expressed to Dr. Seyed Hamed Mirhosseini and Professor Zaidul Islam Sarker, the members of my supervisory committee, for their helpful suggestions throughout my research.

My warmest thanks go also to my parents and husband, Mohsen Kakoienejad. I cannot express how much I would like to say thank you to them for their love and supporting me from day one. They have always been there for me no matter what the situation and I am forever in their debt.

Also, many thanks to my dear friend Navideh Anarjan, I couldn't have completed this research without her support, friendship and encouragement.

I certify that an Examination Committee has met on 29 / 06 / 2012 to conduct the final examination of Mina Tirgar on her Master of Science thesis entitled “Optimization of process conditions and emulsion composition and their effects on the emulsion and encapsulation properties of spray-dried fish oil powder.”

Members of the Thesis Examination Committee were as follows:

Name of Chairman, PhD

Title

Name of Faculty

Universiti Putra Malaysia

(Chairman)

Name of Examiner 1, PhD

Title

Name of Faculty

Universiti Putra Malaysia

(Internal Examiner)

Name of Examiner 2, PhD

Title

Name of Faculty

Universiti Putra Malaysia

(Internal Examiner)

Name of External Examiner, PhD

Title

Name of Department and/ or Faculty

Name of Organization (University / Institute)

(External Examiner)

SIEW HENG FONG, PhD

Professor and Deputy Dean

School of Graduate Studies

Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

Jinap Selamat, PhD

Professor
Faculty of Food Science and Technology
Universiti Putra Malaysia
(Chairman)

Seyed Hamed Mirhosseini, PhD

Lecturer
Faculty of Food Science and Technology
Universiti Putra Malaysia
(Member)

Zaidul Islam Sarker, PhD

Professor
Faculty of Pharmacy
International Islamic University Malaysia
(Member)

BUJANG BIN KIM HUAT, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or other institutions.



TABLE OF CONTENTS

DEDICATION	Page
ABSTRACT	ii
ABSTRAK	v
ACKNOWLEDGEMENTS	viii
APPROVAL	ix
DECLARATION	xi
LIST OF TABLES	xvi
LIST OF FIGURES	xix
LIST OF APPENDICES	xxi
LIST OF ABBRIVIATIONS	xxii
CHAPTER	
1 INTRODUCTION	1
1.1 Background of study	1
1.2 Problem Statement	2
1.3 Objectives	3
1.4 Hypothesis	4
2 LITRATURE REWIEW	5
2.1 Fish oil	5
2.2 Microencapsulation	8
2.2.1 Reasons why the food industry applies microencapsulation	10
2.2.2 Applications of microencapsulation in the food industry	11
2.2.3 Wall material and required properties for encapsulation	13
2.2.4 Properties of the wall material used for encapsulating by spray-drying	16
2.2.5 Carbohydrate for encapsulation by spray-drying	18
2.2.5.1 Maltodextrin	19
2.2.5.2 Arabic gum	20
2.2.5.3 Methylcellulose	21
2.2.6 Other ingredients as wall material for encapsulation	21
2.2.7 Core materials of encapsulation	24
2.2.7.1 Polyunsaturated fatty acids	24

2.2.7.2	Other food constituents for encapsulation	25
2.3	Conventional technologies for encapsulation	28
2.3.1	Physical methods for encapsulation	29
2.3.1.1	Spray-drying	29
2.3.1.2	Other physical drying methods for encapsulation	30
2.3.2	Chemical methods for encapsulation	32
2.3.3	Physicochemical methods for encapsulation	33
2.4	Microencapsulation process steps by spray-drying	36
2.4.1	Atomization	38
2.4.2	Contact of droplet with hot air	39
2.4.3	Evaporation of droplet water	39
2.4.4	Dried product separation	41
2.4.5	Operating conditions of encapsulation procedure	41
2.5	Encapsulation of the fish oil	44
3	SCREENING OF COATING MATERIALS AND OPTIMIZATION OF EMULSION FORMULATION FOR MICROENCAPSULATION OF FISH OIL POWDER	50
3.1	Introduction	50
3.2	Materials and methods	52
3.2.1	Chemicals and materials	52
3.2.2	Sample preparation	53
3.2.3	Homogenization	53
3.2.4	Spray-drying	54
3.3	Analytical methods	54
3.3.1	Emulsion droplet size analysis	54
3.3.2	Centrifugal stability of the emulsion	55
3.3.3	Surface oil content of the powder	56
3.3.4	Emulsion stability (creaming test)	56
3.3.5	Surface mean diameter of the powder	57
3.3.6	Moisture content of the powder	57
3.4	Experimental design and statistical analysis	58
3.5	Optimization and validation procedure	63
3.6	Results and discussion	64
3.6.1	Fitting the response models for screening of the best coating material	64

3.6.1.1	Emulsion droplet size analysis	66
3.6.1.2	Centrifugal stability of the emulsion	70
3.6.1.3	Surface oil of the powder	72
3.6.2	Optimization procedure of the coating material	74
3.6.3	Verification of the reduced models for screening of the best coating material	75
3.6.4	Fitting the response models for optimization of fish oil emulsion formulation	77
3.6.4.1	Emulsion droplet size analysis	80
3.6.4.2	Centrifugal stability of the emulsion	82
3.6.4.3	Creaming stability of the emulsion	84
3.6.4.4	Surface mean diameter of the powder	85
3.6.4.5	Moisture content of the powder	87
3.6.5	Optimization procedure of the fish oil emulsion formulation	88
3.6.6	Verification of the reduced models for optimization of the fish oil emulsion	89
3.7	Conclusion	91
4	PROCESS OPTIMIZATION OF ENCAPSULATED FISH OIL POWDER USING THE SPRAY-DRYING METHOD	93
4.1	Introduction	93
4.2	Materials and methods	95
4.2.1	Chemicals and materials	95
4.2.2	Sample preparation	95
4.2.3	Homogenization	96
4.2.4	Spray-drying	96
4.3	Analytical methods	97
4.3.1	Average particle size and uniformity analysis of powder	97
4.3.2	Total oil content of the encapsulated fish oil powder	97
4.3.3	Surface oil content of the encapsulated fish oil powder	98
4.3.4	Microencapsulation efficiency of the procedure (MEE %)	98
4.3.5	Moisture content of the encapsulated fish oil powder	98
4.3.6	Color measurement of the encapsulated fish oil powder	99
4.3.7	Particle morphology of the encapsulated fish oil powder	99
4.3.8	Solubility index of the encapsulated fish oil powder	99
4.4	Experimental design and statistical analysis	100

4.5	Optimization and validation procedures	102
4.6	Results and discussion	102
4.6.1	Fitting the response models for optimization of the processing conditions	102
4.6.1.1	Average particle size and uniformity analysis of the powder	105
4.6.1.2	Moisture content of the encapsulated fish oil powder	108
4.6.1.3	Total oil content of the encapsulated fish oil powder	109
4.6.1.4	Surface oil content of the encapsulated fish oil powder	112
4.6.1.5	Microencapsulation efficiency of the procedure (MEE %)	115
4.6.1.6	Color analysis of the encapsulated fish oil powder	118
4.6.2	Optimization procedure of processing conditions	121
4.6.3	Verification of the reduced models for optimization of processing conditions	122
4.6.4	Particle morphology of the optimized encapsulated fish oil powder	125
4.6.5	Solubility index of the optimized encapsulated fish oil powder	126
4.7	Conclusion	126
5	GENERAL CONCLUSION AND RECOMMENDATION FOR FUTURE RESEARCH	128
	REFERENCES	131
	BIODATA OF STUDENT	161
	LIST OF PUBLICATIONS	162